

Oscillatory spin relaxation rates in quantum dots

C. F. Destefani and Sergio E. Ulloa

Department of Physics and Astronomy and Nanoscale and Quantum
Phenomena Institute, Ohio University, Athens, Ohio 45701-2979

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Phonon-induced spin relaxation rates in quantum dots are studied as function of in-plane and perpendicular magnetic fields, temperature and electric field, for different dot sizes. We consider Rashba and Dresselhaus spin-orbit mixing in different materials, and show how Zeeman sublevels can relax via piezoelectric and deformation potential coupling to acoustic phonons. We find that strong lateral and vertical confinements may induce minima in the rates at particular values of the magnetic field, where spin relaxation times can reach even seconds. We obtain good agreement with experimental findings in GaAs quantum dots.

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Since the proposal of a qubit based on the electron spin of quantum dots (QDs),¹ much work has been done to understand the processes that may cause their relaxation, since long coherence times are required. One of those processes is related to the phonon-induced spin-flip rates of Zeeman sublevels in QDs in magnetic fields, where the spin purity of the levels is broken by the spin-orbit (SO) interaction. A recent experiment² has shown that spin relaxation time has a lower bound of 50 μ s at an in-plane field of 7.5 T in a GaAs QD defined in a 2DEG, while theoretical work³ has given 5 μ s at the same QD parameters. In general, SO effects have been considered via perturbation theory,^{4,5} although exact treatments have also been presented.^{6,7,8} The perturbative approach, which includes only a few states, has been called into question by the demonstration that a larger basis is needed in order to achieve convergence even for the lowest QD states when the QD vertical width is narrow,⁷ where a complex interplay between different energy scales is present.⁹ The influence of high SO coupling on the QD effective g -factor has been recently addressed.¹⁰

Using exact diagonalization, our goal is to provide a bridge between the different approaches used so far, so that their range of validity can be well established in terms of materials (wide/narrow-gap), QD sizes, and directions of the magnetic field. For GaAs we show that the perturbative approach is indeed satisfactory for in-plane fields *for any QD confinement*, where good agreement is found with calculation^{3,6} and experiment,² and where the *piezoelectric* potential is shown to dominate. If the field is perpendicular, however, we find that the perturbative approach⁴ indeed fails, giving relaxation rates orders of magnitude longer than our results *if the z -confinement is strong*.⁷ For higher perpendicular fields, we show that *minima* are found in the rates depending on the QD features. For InSb the *oscillatory rates* are found for both in-plane and perpendicular fields and strong confinement. We find that the *deformation* potential dominates the rates, so that previous results⁶ under in-plane fields not including such potential yielded rates orders of magnitude smaller than ours. We show that the oscillatory

behavior is caused by the nature of the z -confinement, as well as by the complex interplay between the physical scales playing a role in the system: Zeeman and SO energies, as well as QD lateral and vertical sizes.

The QD is defined by an in-plane parabolic confinement, $V(\rho) = m\omega_0^2\rho^2/2$, where m ($\omega_0 = E_0/\hbar$) is the electronic effective mass (confinement frequency); the QD lateral length is $l_0 = \sqrt{\hbar/(m\omega_0)}$. The vertical confinement $V(z)$ is strong enough so that only the state in the first quantum well subband is relevant, and its function is $\varphi_z(z) = \sqrt{2/z_0} \sin(\pi z/z_0)$ if a hard wall is assumed, z_0 being the QD vertical well thickness. In a magnetic field \mathbf{B} , the unperturbed Hamiltonian, $H_0 = \hbar^2\mathbf{k}^2/2m + V(\rho) + H_Z$, has the well-known Fock-Darwin (FD) solution,¹¹ where $H_Z = g_0\mu_B\mathbf{B} \cdot \boldsymbol{\sigma}/2$ is the Zeeman term. We include all SO terms in 2D zincblende QDs, namely Rashba¹² and Dresselhaus¹³ interactions. The former is due to surface inversion asymmetry (SIA) induced by the 2D confinement, while the latter is caused by the bulk inversion asymmetry (BIA) present in zincblende structures. The SIA Hamiltonian is $H_{SIA} = \alpha\boldsymbol{\sigma} \cdot \nabla V(\rho, z) \times \mathbf{k}$, while the BIA is $H_{BIA} = \gamma[\sigma_x k_x (k_y^2 - k_z^2) + \sigma_y k_y (k_z^2 - k_x^2) + \sigma_z k_z (k_x^2 - k_y^2)]$, with coupling constants α and γ . The z -confinement yields the electric field dV/dz in the SIA Hamiltonian as well as the momentum average $\langle k_z^2 \rangle = (\pi/z_0)^2$ in the BIA terms. The full QD Hamiltonian is then $H = H_0 + H_{SIA} + H_{BIA}$, which is diagonalized in a basis containing 110 FD states. Details about the derivation of terms in H , as well as their selection rules for (anti)crossings in the QD energy spectrum, are found elsewhere.^{8,10}

We calculate spin relaxation rates between the two lowest QD Zeeman sublevels caused by piezoelectric and deformation acoustic phonons via Fermi's Golden Rule: $\Gamma_{fi} = 2\pi/\hbar \sum_{j, \mathbf{Q}} |\gamma_{fi}(\mathbf{q})|^2 |Z(q_z)|^2 |M_j(\mathbf{Q})|^2 (n_Q + 1) \delta(\Delta E + \hbar c_j Q)$, where the sum is over the emitted phonon modes j ($j = LA, TA1, TA2$) with momentum $\mathbf{Q} = (\mathbf{q}, q_z)$. The term $Z(q_z) = \langle \varphi_z | e^{iq_z z} | \varphi_z \rangle$ ($\gamma_{fi}(\mathbf{q}) = \langle f | e^{i\mathbf{q} \cdot \mathbf{r}} | i \rangle$) is the form factor perpendicular (parallel) to the 2D-plane (position is $\mathbf{R} = (\mathbf{r}, z)$), while n_Q is the phonon distribution with energy $\hbar c_j Q$; energies

$\Delta E = \varepsilon_f - \varepsilon_i$ and states $|i\rangle, |f\rangle$ are obtained via diagonalization of the total H , so that the SO mixing is fully taken into account. The element $M_j(\mathbf{Q}) = \Lambda_j(\mathbf{Q}) + i\Xi_j(\mathbf{Q})$ includes both piezoelectric Λ_j and deformation Ξ_j potentials; in zincblende structures¹⁴ and in cylindrical coordinates, they become $\Xi_{LA}(\mathbf{Q}) = \Xi_0 A_{LA} \sqrt{Q}$ (only LA is present for Ξ_j), $\Lambda_{LA}(\mathbf{Q}) = 3\Lambda_0 A_{LA} \sin(2\theta) q^2 q_z / 2Q^{7/2}$, $\Lambda_{TA1}(\mathbf{Q}) = \Lambda_0 A_{TA} \cos(2\theta) q q_z / Q^{5/2}$, and $\Lambda_{TA2}(\mathbf{Q}) = \Lambda_0 A_{TA} \sin(2\theta) (2q_z^2 / q^2 - 1) q^3 / 2Q^{7/2}$ (both $TA1$ and $TA2$ modes are compacted as a single TA mode for Λ_j), where $A_j = \sqrt{\hbar(2N_0 V c_j)^{-1}}$ and $\Lambda_0 = 4\pi e \hbar_{14} / \kappa$. The bulk phonon constants are Ξ_0 and $e \hbar_{14}$, c_j are the sound velocities ($c_{TA1} = c_{TA2} = c_{TA} \neq c_{LA}$), κ is the dielectric constant, and N_0 is the electron density. The triple space integration ($[r, \phi_r, z]$) yields an analytical solution,⁷ and we were able to do two ($[\phi_q, q_z]$) out of the momentum integration, leaving a numerical integral only in q . For a later use, the only z_0 -dependence in this remaining integral in Γ_{fi} reads $F_j(z_0) = (d_j z_0 - (d_j z_0)^3 / \pi^2)^{-2} \sin^2(d_j z_0)$, where $d_j = \sqrt{(\Delta E / \hbar c_j)^2 - q^2 / 2}$; q runs from 0 to $\Delta E / \hbar c_j$, while $F_j(z_0)$ is multiplied by polynomials and exponentials in q in the total Γ_{fi} . No approximation is needed in our derivation of Γ_{fi} , so that the 3D nature of the phonon is taken into account and the full form factor $e^{i\mathbf{Q} \cdot \mathbf{R}}$ is used.

Panel A of Fig. 1 shows the phonon-induced spin-flip rates as a function of in-plane field, B_{\parallel} , for different values of E_0 (solid lines) for GaAs QDs.¹⁵ Larger E_0 (smaller QDs) present smaller spin-flip rates, i.e., longer relaxation times. This happens because as E_0 increases, the orbital levels become more separated and then the SO coupling becomes relatively less important. A strong dependence of the rates with B_{\parallel} and E_0 is clear. For example, at $B_{\parallel} = 2$ T, one gets $2.5 \times 10^2 \text{ s}^{-1}$ at $E_0 = 0.7$ meV and $2.5 \times 10^{-1} \text{ s}^{-1}$ at $E_0 = 5.0$ meV; at $B_{\parallel} = 15$ T the rate goes from $5.2 \times 10^6 \text{ s}^{-1}$ to $6.9 \times 10^4 \text{ s}^{-1}$ for those same values of E_0 . Finite temperature (dashed-dotted line for $T = 12$ K) enhances the rates by about one order of magnitude due to the enhancement of the phonon population. Larger wells (dotted line for $z_0 = 100$ Å) have rates about one order of magnitude smaller because the BIA term ($\langle k_z^2 \rangle \approx 1/z_0^2$) decreases. The same T and z_0 behavior is found for any E_0 . Larger electric fields (dashed line for $dV/dz = -38 \text{ meV/\AA}$) have little effect on the rates for large E_0 , but at smaller E_0 the rates decrease by one order of magnitude; this means that by increasing the SIA term so that it equals the BIA coupling, the rates can decrease even though the spin mixing gets stronger. For GaAs QDs under *in-plane* fields, we find that the *TA piezoelectric* coupling dominates at *low fields* ($\lesssim 14$ T for $E_0 = 5$ meV), while at high fields the deformation potential takes over (symbols in Fig. 1A).

Panels B and C of Fig. 1 respectively show the spin expectation value $\langle \sigma_z \rangle$ of the Zeeman sublevels and their energy splitting ΔE . Smaller E_0 values show higher SO-induced spin mixing, so that $\langle \sigma_z \rangle$ deviates from the pure +1 (−1) value for the ground (first excited) state, and

FIG. 1: (A). Zeeman sublevel spin-flip rates for GaAs QDs¹⁵ under in-plane magnetic field for different QD confinements (solid lines). Dashed-dotted, dotted, and dashed lines respectively show influence of higher temperature, well width, and electric field on the 3.5 meV QD. Symbols show isolated contributions from the distinct phonon mechanisms for the 5.0 meV QD; TA piezoelectric dominates at low fields. (B). Spin average values of the Zeeman sublevels after diagonalization of H . Squares (circles) refer to ground (first excited) state. (C). Energy splitting of Zeeman sublevels after diagonalization of H . Dashed-dotted lines show results without SO coupling. Color scheme in panels B and C follow from panel A.

FIG. 2: Same as Fig. 1, but for GaAs QDs under perpendicular magnetic field. Vertical lines in panel B show the field where a sublevel crossing occurs and the normal spin character of the states is restored. Arrows in panel C indicate where suppression of SO coupling is induced by the magnetic field, which is at the origin of the respective minima in panel A.

ΔE deviates from the pure $g_0 \mu_B B$ value of a QD without SO coupling. Notice at the 3.5 meV QD how a larger well (stronger field) decreases (increases) the SO-induced spin mixing by reducing (enhancing) the importance of the BIA (SIA) coupling. For comparison, the experimental lower bound for the spin-flip time is $50 \mu\text{s}$ at $B_{\parallel} = 7.5$ T for a 1.1 meV QD,² while from panel A we find $5 \mu\text{s}$. As discussed above, this time is increased by one order of magnitude in a larger z -well, which appears to be a better match for the experimental conditions. A $5 \mu\text{s}$ value has also been found from a perturbative formulation,³ and our results also agree with a different model for the vertical confinement.⁶ One can then affirm that the perturbative approach⁴ is indeed enough for GaAs in *in-plane* fields at *any* QD sizes, and that the form of the z -confinement has little effect on the rates.

In Fig. 2 we do the same analysis for GaAs QDs¹⁵ in a perpendicular field, B_{\perp} . The most striking feature is the appearance of minima in the rates shown in panel A. The E_0 -dependent minima ($12 \leq B_{\perp} \leq 16$ T) are due to the *vanishing* ΔE , as shown in panel C. At such values of B_{\perp} , a level crossing of Zeeman sublevels results in a sudden *spin-flip*, as verified by the vertical lines of panel B for the 0.7 and 3.5 meV QDs. To understand the minima at low fields ($6 \leq B_{\perp} \leq 8$ T) we have to be mindful of the sine argument in $F_j(z_0)$, which may induce a minimum in the rate at a particular value of the field, due to the interplay of energy and length scales in the problem.⁹ This behavior does not produce minima for in-plane field on GaAs QDs (Fig. 1). We find that those three low-field minima in Fig. 2A occur at B_{\perp} values where a *magnetic field-induced cancellation* of the SO influence is produced on the respective ΔE values, as indicated by the arrows in panel C: below (above) such E_0 -dependent values of B_{\perp} – where ΔE recovers its Zeeman value of $g_0 \mu_B B$ – the SO coupling increases (decreases) the Zeeman splitting as compared to the QD without SO.

FIG. 3: InSb QDs¹⁷ under in-plane magnetic field. The confinements are such that the same QD size-range used for GaAs is covered. In panel A, the 15.0 (20.0) meV QD is chosen for the study of different QD parameters (distinct phonon mechanisms); dashed arrows show that both LA modes have the same number of rate minima and happen at the same fields.

Notice that the two smallest confinements do not show the low-field rate minima in Fig. 2A. This is due to the lowest sublevels acquiring the same spin (between 3 and 7 T at $E_0 = 0.7$ meV in panel B), so that the rate decreases monotonically at that field-range. These features have strong influence on QD effective g -factors.¹⁰ Like in the in-plane field problem, higher temperatures ($T = 12$ K) enhance the rates by one order of magnitude, and the dominating phonon mechanism (symbols) is the TA piezoelectric coupling, but now at *any* field. To confirm the intricate interplay between all QD energy scales, a larger well ($z_0 = 100$ Å) or a stronger field ($dV/dz = -38$ meV/Å) *removes* the rate minima, as shown in panel A for the 3.5 meV QD. Our results agree with available calculations⁷ at small fields ($B_\perp \leq 1$ T), so that we can confirm that the perturbative approach is *not adequate* when dealing with SO effects in QDs in a *perpendicular* field, even if the material is GaAs, *if the vertical confinement is strong enough*.¹⁶ From Figs. 1 and 2 the rates at small fields ($\lesssim 2$ T) have the same behavior under B_\parallel and B_\perp , namely, the smaller E_0 the larger the rate; however, they are much larger under B_\perp . The rate maximum (at 2.5 T for the 1.1 meV QD) is due to an anticrossing involving the first and second QD excited states.¹⁰

Figure 3 shows results for InSb QDs¹⁷ in in-plane fields. In contrast to GaAs (Fig. 1), minima in the rates are visible under B_\parallel for the highest E_0 values. At small fields ($\lesssim 2$ T), a monotonic rate drop is again observed as E_0 increases. As shown for the 20 meV QD, the *LA deformation potential* is the dominant phonon mechanism for InSb at *any* field (and E_0); this seems to be the case for any narrow-gap material.¹⁸ It is worth mentioning that, contrary to GaAs in Fig. 2A where a unique low-field minimum is present for a given E_0 , the number of rate minima in InSb increases with E_0 . Also, notice that both LA modes have the same number of minima at the same B_\parallel values (dashed arrows in panel A), while a larger number of minima in the TA mode occurs at different fields; this happens since $c_{LA} > c_{TA}$, so that the sine argument in $F_j(z_0)$ is smaller in the LA mode. Panel B shows that Zeeman sublevels have normal spin character, where ground (excited) state is predominantly spin-up (spin-down) at low fields. Finite temperature ($T = 12$ K) has no appreciable effect on the rates, a reflection of the much larger energy scales here. The energy scale interplay results at higher field ($dV/dz = -2$ meV/Å) in slightly smaller (larger) ΔE (rates), and rate minimum

shifts to higher fields. On the other hand, a larger well ($z_0 = 100$ Å) produces a higher ΔE , so that the spin-flip rate is suppressed by orders of magnitude and its num-

FIG. 4: Same as Fig. 3, but for InSb QDs under perpendicular magnetic field. The only minimum of the 3.0 meV QD around 1 T in panel A indicates a sublevel crossing, as shown by the vertical line in panel B and the vanishing of ΔE in panel C. The color scheme is the same in all panels and figures.

ber of minima increases; notice in panel B that the two lowest levels acquire the same spin at $\simeq 8$ T for such z_0 , so that the oscillatory rate is well defined until that field. Previous calculations⁶ in InSb did not include the deformation potential, and yielded unrealistically low rates.

For InSb QDs in perpendicular fields, we see from panel B in Fig. 4 that all spin-flip rates in panel A are meaningful only for $B_\perp < 8$ T, a field where both Zeeman sublevels acquire the same spin. This happens earlier for small E_0 as shown by the 3.0 meV QD, whose rates are well defined only for $B_\perp < 2$ T; the vertical line around 1 T in panel B for this E_0 indicates a spin-flip, accompanied by the vanishing ΔE in panel C, the rate minimum in panel A, and a sign change of the QD g -factor.¹⁰ Larger values of E_0 present normal spin behavior. Spin-flip rates in panel A do *not* show the monotonic behavior seen for small fields in Fig. 3. Like in the B_\parallel case, a finite temperature ($T = 12$ K) does not show visible influence on the rates under B_\perp , and the deformation potential dominates at any field. A higher electric field ($dV/dz = -2$ meV/Å) slightly increases the rate; a larger well ($z_0 = 100$ Å) *introduces* an oscillatory rate orders of magnitude smaller. We emphasize that the perturbative approach finds *no use* in InSb QDs because of the inherent higher SO coupling. It is worth mention that oscillatory rates have also been found in GaAs QDs under B_\perp by considering *momentum* relaxation from an excited orbital level to the ground state,¹⁹ as well as in coupled GaAs QDs.²⁰

Even though both QD materials show oscillatory spin-flip rates – with B_\perp for GaAs and with both B_\parallel and B_\perp for InSb – their origin is slightly different. Minima come from the nature of the z -confinement, and the field where they occur depend on the lateral size l_0 . The SO coupling mixes spins and alters splitting of sublevels in distinct ways according to field-direction and QD material, so that spin relaxation can be induced by piezoelectric (wide-gap) and deformation (narrow-gap) phonons. Our calculations reveal the rich interplay between all relevant energy scales in QDs. The external field opens channels (at the rate minima) where long spin relaxation times ($\simeq 1$ s) may be reached, so that the spin coherence required for quantum computing could be improved.

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- ¹⁶ $\langle k_z^2 \rangle = (\pi/z_0)^2$ for an infinite well. If a Gaussian well is considered, one has $\langle k_z^2 \rangle = 1/z_0^2$, which yields a BIA term a factor of 10 smaller than used in this work.
- ¹⁷ In InSb: $m = 0.014 m_0$, $g_0 = -51$, $\kappa = 16.5$, $\alpha = 500 \text{ \AA}^2$, $\gamma = 160 \text{ eV \AA}^3$, $\Xi_0 = 7 \text{ eV}$, $eh_{14} = 0.061 \text{ eV/\AA}$, $N_0 = 5.77 \times 10^{-27} \text{ Kg/\AA}^3$, $c_{LA} = 3.40 \times 10^{13} \text{ \AA/s}$, and $c_{TA} = 2.29 \times 10^{13} \text{ \AA/s}$; also, $z_0 = 40 \text{ \AA}$, $dV/dz = -0.5 \text{ meV/\AA}$, and $T = 0 \text{ K}$ if no other numbers are specified.
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